

English language abstract). Applicant requests withdrawal of these rejections in view of the following comments.

This invention is directed to a method for increasing ethylene yield during conversion of oxygenates to olefins. In particular, the ethylene content is increased by: (1) recycle of a selective hydrocarbon product stream; or (2) the use of a certain type of second reactor system to further modify the selective hydrocarbon product stream. The method uses a small or medium pore non-zeolitic (e.g., SAPO) molecular sieve catalyst as a primary contact catalyst in the conversion process.

The UK reference is primarily directed to a method of making propylene and a minor proportion of butenes by contacting methanol and/or dimethyl ether with a dealuminated mordenite catalyst. The mordenite catalyst is taught to be known to have a good selectivity for olefin production (page 1, lines 48-54). As is known in the art, mordenite molecular sieve catalysts are large pore zeolite catalysts. See *Atlas of Zeolite Structure Types*, Meier et. al., Butterworth-Heinemann, 1992 (copy attached). According to the UK reference, the dealuminated zeolite catalyst can be used to improve propylene yield using specific operating conditions (page 1, lines 55-56). In particular, lines 24-28 of page 2 indicate that it is preferred to recycle a C_4^+ olefin stream in order to increase the propylene yield.

Applicant's claimed invention differs from the UK reference in that applicant uses a small or medium pore non-zeolite catalyst in the reaction process, not a large pore zeolite. Applicant has found that contacting a heavy hydrocarbon product stream and an oxygenate feed with that type of catalyst increases light olefin yield as a whole, with a tendency to be richer in ethylene content. There is nothing in the UK reference to suggest the use of such a catalyst or that recycling a heavy hydrocarbon would produce a light olefin stream rich in ethylene. In fact, the UK reference seeks to increase the propylene content. Additional ethylene production is not favored. The fact that the UK reference uses a large pore zeolite molecular sieve tends to further

suggest the desirability of propylene production over ethylene, since propylene is a larger molecule than ethylene.

The Kaiser '243 patent discloses making light olefin products by contacting aliphatic hetero compounds such as methanol and dimethyl ether with a non-zeolitic, SAPO molecular sieve catalyst. Kaiser's definition of a light olefin product, however, is a C₂-C₄ olefin (col. 5, lines 52-53), not a C₂-C₃ stream as defined by applicant (see page 1, line 13, of the specification). The C₂-C₄ olefin comprises a major portion of the product, while a minor portion of the product is methane and aromatics (col. 5, lines 53-56). The '243 patent does not discuss separation of components in the product stream, recycle of product components, or successive conversion of heavy hydrocarbon product. There is also no teaching regarding any desire to increase ethylene or propylene production specifically, as the patent is primarily concerned with the general increase of C₂-C₄ olefin content as a whole. Combining the teaching of the Kaiser '243 patent with that of the UK patent does not change the focus of the UK patent away from producing propylene. Changing the UK mordenite catalyst to a SAPO catalyst would essentially result in doing away with the entire teaching of the UK reference to the extent that increased production of propylene would be lost. Therefore, the combination of Kaiser '243 and the UK patent not only does not suggest applicant's claimed invention, it would destroy the concept behind the UK disclosure.

The Kaiser '001 patent describes the interconversion of small olefins with non-zeolitic molecular sieves. Lines 55-63 of column 2 states that the definition of "olefin conversion" includes the conversion of butenes to ethylene and butenes to propylene. According to the data shown in the '001 patent, contacting 1-butene with a non-zeolitic (SAPO-34) catalyst (Tables III and VIII) results in only a slight conversion of 1-butene to ethylene and propylene. In fact, the data show that the predominant conversion of 1-butene is to propylene. Although there is some conversion to C₅⁺ (i.e., C₅ + C₆) components, the percent conversion to those components is very close to that of ethylene.

Applicant's data in the Table in Example 1 confirm the general findings of the Kaiser '001 patent. That is, the data in applicant's Example 1 show that the conversion of 1-butene is very low (34.23%), and the predominant product is propylene and C_5^+ components. In comparison, applicant's Example 1 further shows that methanol is predominantly converted to ethylene.

The data in Example 1 further show that when methanol and 1-butene are mixed together and contacted with a silicoaluminophosphate molecular sieve, the result is a product which is high in light olefin and rich in ethylene -- not merely the average of converting separate butene and methanol streams. This demonstrates that combining a heavy hydrocarbon product fraction with oxygenate in the feed stream, and contacting the combined feed with a small or medium pore non-zeolitic catalyst, results in a significant shift to ethylene content while achieving a high light olefin content. In addition, such a method results in very little C_5^+ by-product. The amount of C_5^+ by-product that is formed is actually comparable to that of a methanol only feed; and the conversion of the methanol + 1-butene streams remains very high. Such a method is not suggested by any combination of the cited references. The shift to ethylene formation is not suggested by any of the cited references taken alone or in combination. In fact, the Kaiser '001 reference suggests that recycle of heavy hydrocarbon (e.g., butenes) would substantially increase the formation of propylene and C_5^+ components. This clearly teaches away from applicant's claimed invention.

The English language abstract of the German reference discloses a process of preparing olefins by contacting methanol with a ZSM type of zeolite catalyst. The method disclosed in this reference is very similar to the method of the UK reference. The difference in the UK reference and the German reference is that the catalyst in the German reference is defined as a ZSM type catalyst which is generally a medium pore type of zeolite molecular sieve with a high degree of similarity to the dealuminated catalyst of the UK reference. These types of sieves typically tend to provide larger products such as propylene, rather than the smaller ethylene product. In any

event, there is nothing stated in the German reference that would suggest modifying any of the other cited references. If anything, combining the teachings of the UK reference with the German reference would suggest that recycle of a heavy by-product would be just as beneficial for increasing propylene yield using a ZSM catalyst as for a non-zeolite catalyst. Accordingly, none of the cited references taken alone or in combination suggests applicant's claimed invention.

Having demonstrated that applicant's claimed invention is not disclosed or suggested by the cited references, this application is in condition for allowance. Accordingly, early and favorable reconsideration is requested.

Attached hereto is a marked-up version of the changes made to the specification and claims by the current amendment. The attached page is captioned "**Version with markings to show changes made**".

In the unlikely event that the transmittal letter is separated from this document and the Patent Office determines that an extension and/or other relief is required, applicant petitions for any required relief including extensions of time and authorizes the Commissioner to charge the cost of such petitions and/or other fees due in connection with the filing of this document to **Deposit Account No. 03-1952** referencing docket no. 357972004600. However, the Commissioner is not authorized to charge the cost of the issue fee to the Deposit Account.

Respectfully submitted,

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By:



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ATLAS OF ZEOLITE STRUCTURE TYPES

W.M. Meier and D.H. Olson



Third Revised Edition
1992

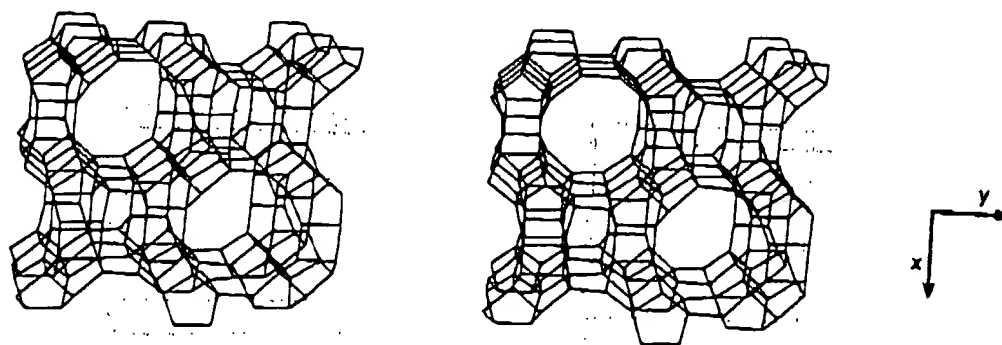
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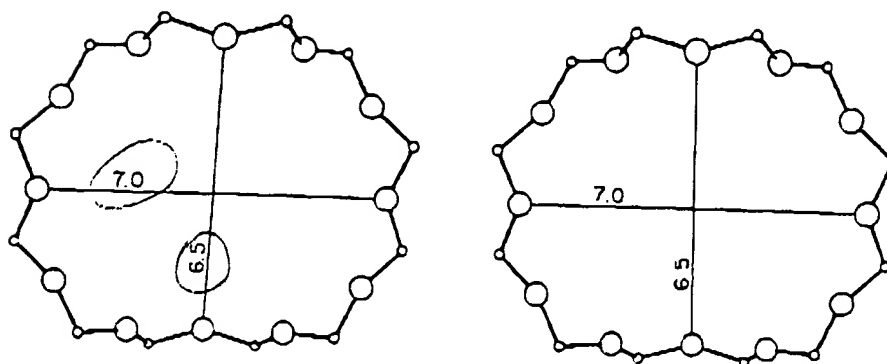
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$$16 T_{1,2}(1) \cdot 8 T_{3,4}(m)$$

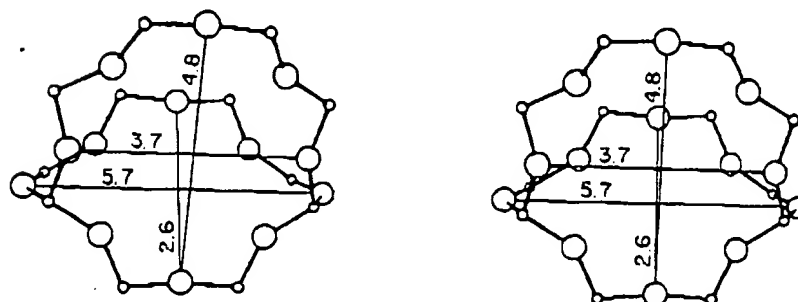
MOR



framework viewed along [001]



12-ring viewed along [001]



complex of 8-rings viewed along [010]

MOR

MORDENITE

Cmcm

16

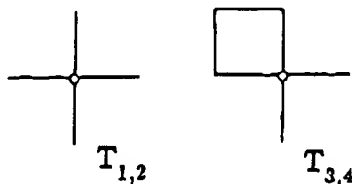
Secondary building
units:

5-1

Framework density:

17.2 T/1000 Å³

Loop configuration
of T-atoms:



Coordination sequences:

T₁(16) 4 12 22 38 60 88 115 155 204 242
 T₂(16) 4 12 20 37 64 87 114 154 198 241
 T₃(8) 4 11 24 39 54 86 126 156 195 242
 T₄(8) 4 11 24 39 60 92 122 148 195 250

Channels:

[001] 12 6.5 x 7.0* ↔ [010] 8 2.6 x 5.7*

Fault planes:

(010), (100)

Type material:

Mordenite Na₈[Al₈Si₄₀O₉₆] · 24 H₂O
 orthorhombic, Cmcm, a=18.1, b=20.5, c=7.5 Å⁽¹⁾

Isotypic framework
structures:

Na-D⁽²⁾
 Ca-Q⁽³⁾
 Zeolon

Large port mordenite⁽⁴⁾
~~LZ-211⁽⁵⁾~~

Alternate designations:

Ptilolite Flokite (discredited)
 Arduinite (discredited)

References:

- (1) W. M. Meier, Z. Kristallogr. 115, 439 (1961).
- (2) R. M. Barrer and E. A. D. White, J. Chem. Soc. 1952, 1561 (1952).
- (3) M. Koizumi and R. Roy, J. Geol. 68, 41 (1960).
- (4) L. B. Sand, Molecular Sieves, Soc. of Chem. Industry, London, p. 71 (1968).
- (5) D. W. Breck and G. W. Skeels, US Patent 4,503,023 (1985).